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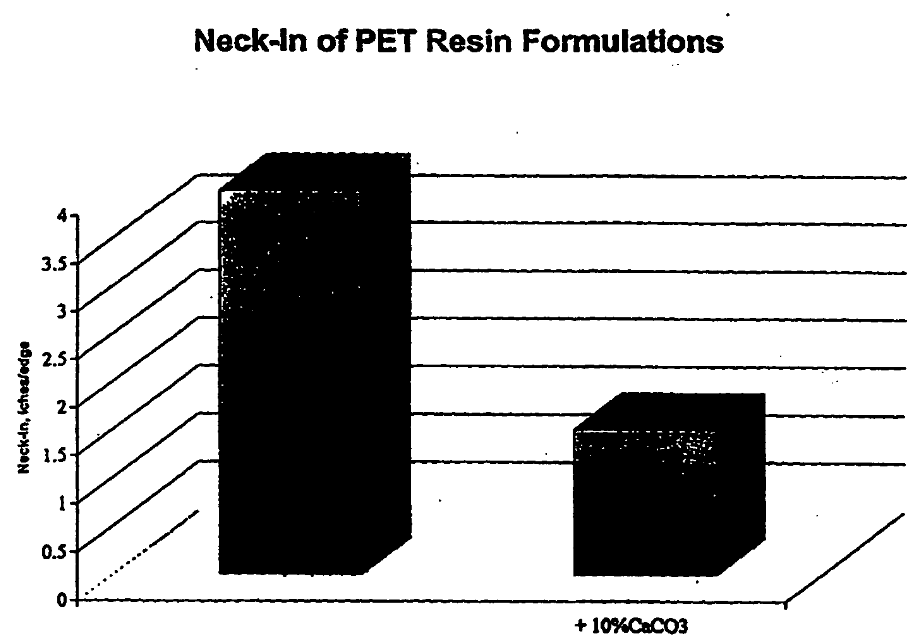
(54) Title: MODIFIED POLYESTER WITH IMPROVED PROCESSABILITY AND ADHESION IN EXTRUSION COATING APPLICATIONS



(57) Abstract

A polyester composition having at least 5 weight % of an inorganic filler exhibits improved processability and adhesion to cellulosic substrates. The composition may be extruded into exceptionally thin films having improved neck-in aspects and used in manufacturing a variety of articles, such as food and beverage containers and is particularly useful in the manufacture of microwaveable cellulosic composite structures.

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MODIFIED POLYESTER WITH IMPROVED PROCESSABILITY AND ADHESION IN EXTRUSION COATING APPLICATIONS

CROSS-REFERENCE TO RELATED APPLICATIONS

This application claims benefit under 35 U.S.C. § 119 to the provisional application having U.S. Serial No. 60/72,019, filed January 21, 1998, the entire disclosure of which is incorporated herein by reference.

FIELD OF THE INVENTION

The present invention relates to polyester compositions having an improved adhesion to a substrate and to an article made therefrom. More particularly, the present invention relates to a polyester composition which includes an inorganic filler and has an improved adhesion to a cellulosic substrate and to an article having at least a portion thereof made from the composition.

BACKGROUND OF THE INVENTION

For many years, polyester homopolymers and copolymers have been used in a wide variety of applications. For example, saturated and unsaturated polyester resins are used in various sheet molding and bulk molding applications where the polyester is mixed with a reinforcing material, such as glass fibers, and fillers in addition to other materials to form such articles as automobile parts. For example, U.S. Patent No. 5,015,295 discloses a polyester molding composition suitable for sheet molding and bulk molding applications. The composition includes a polyester resin, chopped fibers and a Class I calcium carbonate filler. The filler is a finely divided, dry ground micritic Caribbean limestone having a $CaCO_3$ content of at least 95 %.

Polyesters may also be extruded into films and used in electronic applications, such as capacitors and as a substrate for magnetic recording media. For example, U.S. Patent No. 5,674,443 discloses a process for preparing a polyester film having improved surface and mechanical properties by adding calcium carbonate treated with a coupling agent to the polyester. In the process, the surface treated calcium carbonate is added to a portion of the glycol to be used in making the polyester. The resultant slurry is added as a slip agent during the preparation of the polyester, preferably during the transesterification step or immediately prior to the polycondensation step.

U.S. Patent No. 5,747,633 discloses a polyester composition having from 20 weight % to 85.5 weight % of a saturated polyester resin, from 2.5 weight % to 45 weight % of a hydroxyl group-containing resin, such as vinyl alcohol, and from 5 weight % to 50 weight % of an alkaline earth metal carbonate as a filler.

U.S. Patent No. 5,288,784 discloses a polyester composition having vaterite-type calcium carbonate included to improve the slipperiness of the polyester. The vaterite-type calcium carbonate is prepared by carbonating a calcium compound in a mixed medium of alcohol with water.

U.S. Patent No. 4,965,307 discloses a polyester composition having improved slipping and slitting properties when formed into a film suitable for use as a magnetic recording media. The polyester includes not less than 0.005 weight % of calcium carbonate particles having specific size are incorporated into a polyester having crystals of a specified size and orientation.

Yet other applications include food and beverage containers where the polyester is used either alone or in combination with another material to form the container. For example, polyethylene terephthalate (PET) and copolymers thereof, are useful as extrusion coatings due to the material having a good combination of thermal resistance, printability, and organoleptic properties. Polyesters are particularly well suited for coating for various food-packaging applications, such as coatings on frozen food trays, beverage cups, and the like.

However, coating a polyester on a substrate is not without its problems. The low melt strength of polyesters (compared, for example, to low density polyethylene) poses processing problems during extrusion, such as neck-in. Neck-in problems result in non-

uniform coating weights and non-uniform thickness, all of which lead to unsatisfactory products.

Another problem associated with coating a polyester on a substrate is the adhesion of polyesters to both polar and non-polar substrates. Unlike polyethylene resins, polyesters do not readily oxidize in the air gap between the substrate and the coating, and because of their high modulus and rapid rate of solidification, polyesters do not readily penetrate into porous substrates, and particularly cellulosic materials. This problem is typically compensated for by running at relatively heavy coating weights (e.g., 25-50 microns, compared to 12-25 microns using polyethylenes) to achieve a uniform coating with good web stability and acceptable adhesion.

Thus there is a need for a polyester composition that exhibits better neck-in properties during extrusion coating, and provides good adhesion to a substrate at lower coating weights.

SUMMARY OF THE INVENTION

The present inventors have unexpectedly discovered that addition of inorganic fillers, such as calcium carbonate, results in a polyester formulation with dramatically improved processability (higher maximum line speed and thinner minimum coating weight), and good adhesion to a substrate, such as paper, at reduced coating weight.

Briefly, the present invention provides for a polyester composition having a critical amount of an inorganic filler, preferably calcium carbonate. Films derived from the composition of the invention exhibit improved slipperiness in that they have higher film forming line speeds and exhibit improved adhesion to a substrate.

In another aspect of the invention, an article of manufacture is provided having the polyester composition of the invention coated on at least a portion of a substrate. Desirably, the substrate is a cellulosic or cellulose containing material.

It is an object of the present invention to provide a polyester composition having improved neck-in when extruded into a film and adhesion to a substrate.

It is another object of the invention to provide an article of manufacture made from a polyester having at least 5 weight % of an inorganic filler.

It is another object of the invention to provide a composite article having at least a portion of the article coated with a polyester having at least 5 weight % of calcium carbonate.

It is still another object of the invention to teach a process for extrusion coating of polyesters wherein the coating line speed can be increased over non-inorganic filler containing polyester compositions, and wherein good adhesion to the substrate can be obtained for coatings as thin as about 13 microns.

These and other objects, features, and advantages of the present invention will become apparent as reference is made to the following drawings, detailed description, preferred embodiments, and examples.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is an illustration of an extrusion coating and laminating line which may be used for film extruding the polyester composition of the invention.

Figure 2 is a cross-view of the melt curtain. During the extrusion coating process, if the melt curtain becomes unstable the melt curtain will have excessive weaving of the edges.

Figure 3 shows a comparison of the neck-in observed for a polyester composition without addition of inorganic filler (left bar on graph) and with addition of 10% $CaCO_3$ (right bar on graph), at an extrusion coating line speed of 400 fpm.

Figure 4 shows a comparison of the maximum line speed for extrusion coating onto paper of the same polyester compositions as in Figure 1 (left bar is the polyester composition without $CaCO_3$; right bar is the composition according to the present invention).

Figure 5 shows a comparison of the minimum coating weight providing good adhesion to a paper substrate, using the same polyester compositions as in Figure 1 (left bar is the polyester composition without $CaCO_3$; right bar is the composition according to the present invention).

DETAILED DESCRIPTION OF THE INVENTION

The polyester composition according to the present invention, having inorganic filler added, may be a polymer, copolymer, or blends thereof. As used herein, "polyester" refers to polyesters obtained by polycondensation reaction and having residues from starting materials comprising a dicarboxylic acid and a diol. Desirably, the dicarboxylic acid is an aromatic dicarboxylic acid such as terephthalic acid or naphthalene-2,6-dicarboxylic acid or an ester thereof. Desirably, the diol is an aliphatic glycol. Accordingly, polymers that are particularly useful in this process include PET, PEN, and copolyesters thereof containing up to about 50 mole %, and preferably up to about 20 mole % of modifying dibasic acids and/or glycols and blends thereof. Modifying dibasic acids may contain from about 2 to 40 carbon atoms and include isophthalic, succinic, adipic, glutaric, azelaic, sulfoisophthalic, sebacic, fumaric, cis- or trans- 1,4-cyclohexanedicarboxylic, the various isomers of naphthalene dicarboxylic acids and mixtures thereof. Highly useful naphthalene dicarboxylic acids include the 2,6-, 1,4-, 1,5-, or 2,7- isomers but the 1,2-, 1,3-, 1,6-, 1,7-, 1,8-, 2,3-, 2,4-, 2,5-, and/or 2,8- isomers may also be used. The dibasic acids may be used in acid form or as their esters such as the dimethyl esters for example.

Desirably, the aliphatic glycol is ethylene glycol and polymethylene glycols of 2-10 carbon atoms, such as trimethylene glycol, tetramethylene glycol, pentamethylene glycol, hexamethylene glycol, and decamethylene glycol. The glycol component of the polyester may contain from about 97 mole percent to about 35 mole percent ethylene glycol and from about 1.0 mole percent to about 65 mole percent 1,4-cyclohexanedimethanol (CHDM). The glycol component may be further modified with up to 20 mole percent with glycols selected from propylene glycol, 1,3-propanediol, neopentyl glycol, 1,4-butanediol, 1,6-hexanediol, 1,4-cyclohexanediol, diethylene glycol, cis/trans mixtures of 1,4-cyclohexanedimethanol, 2,2,4,4-tetramethyl-1,3-cyclobutanediol and mixtures thereof.

In accordance with one aspect of the invention, the polyester includes at least 5 weight %, preferably from 5 weight % to about 30 weight %, more preferably from about 10 weight % to about 20 weight %, and most preferably from about 10 weight % to

about 15 weight % of an inorganic filler. It is to be understood that the above preferred ranges include all values in between. The preferred inorganic filler is calcium carbonate. The calcium carbonate particles can have an average size of less than about 50 microns, preferably from about 0.01 microns to about 30 microns and most preferably from about 3 microns to about 20 microns. The calcium carbonate can be obtained using methods well known to those skilled in the art and include dry grinding and wet grinding. Moreover, the calcium carbonate may be uncoated but preferably is coated and can be obtained from natural sources or manufactured as described in U.S. patent nos. 4,727,108 and 5,288,784, the entire disclosures of each being incorporated herein by reference.

In adding the calcium carbonate to the polyester, there is no particular limitation as to manner in which it may be included into the polyester. One method is to prepare a master batch by blending the calcium carbonate in an amount in excess of the ranges set forth herein then further blending the resin of this master batch with other polyester to achieve the desired concentration of calcium carbonate in the final product.

In addition to calcium carbonate the polyester may contain other particles in amounts not adversely affecting the advantageous effects of the present invention. For example, the polyester may contain fine particles of amorphous zeolite, anatase titanum dioxide, calcium phosphate, silica, kaolin clay, talc, and the like. Moreover, the polyester composition may contain fine catalyst particles precipitated by the esterification reaction. The amount of these additional materials may be less than about 1 weight % of the total polyester composition.

The polyester composition of the present invention can be blended with a variety of coloring agents, such as, pigments and dyes, reinforcing agents, lubricating agents, plasticizers, leveling agents, surfactants, viscosity-increasing agents, antioxidants, and the like depending upon the application.

In another aspect of the invention, an article of manufacture is made using the polyester composition described above. For example, articles which can be manufactured using the composition of the invention include packaging containers, such as, bottles, cups, tubes, plastic cans, pouches, films which may be used in food applications,

containers for distribution, and particularly preferred are cellulosic composite structures, particularly microwaveable food containers and trays.

In accordance with the practice of the invention, the polyester composition is extruded through a die into a film, which is brought into contact with a substrate to form the article. In a preferred embodiment, the substrate is a cellulosic material or cellulose containing material. For the purpose of description, the terms "cellulosic" and "cellulose containing" are used interchangeably herein. Cellulosic materials can be obtained from vegetable sources, especially wood and may comprise soft wood fibers, hardwood fibers or a mixture thereof and which may be a mechanical, chemimechanical, semichemical or chemical pulp, or may comprise recycled or secondary fibers with or without organic fillers. Other sources of cellulosic materials include cotton, bagasse, esparto, straw, reed, or hemp.

The following examples are for illustration purposes and are not to be a limitation of the invention. In the examples, all parts and percentages are by weight unless otherwise specified.

The equipment used to carry out the examples is described as follows:

A laboratory extrusion coating and laminating line, manufactured by ErWePa Machineinfabrik (now, Egan/Davis-Standard), consists of three extruders: Extruder A is a 2.5" diameter, 24:1 L/D, single-flighted design, with a compression ratio of 3:1; Extruders B and C are 2" diameter, with screws comparable to Extruder A. The three extruders feed into a standard feed block and into the die, which in this case is an internally deckled "bead-reduction" design by the Cloeren Company. This set-up is illustrated in Figure 1.

The laminator portion of the line included primary and secondary substrate unwind stations, corona and flame pre-treating stations, a coating/nip station equipped with a water-cooled metal chill roll, a rubber nip roll, and a re-wind station. A side view of the line is depicted in Fig. 1. A cross-view of the melt curtain is depicted in Figure 2. During the extrusion coating process, if the melt curtain becomes unstable, as evidenced by excessive weaving of the edges, then one is no longer able to produce an acceptable product. To one skilled in the art of extrusion coating, if the neck-in increases as the line speed increases, the resin is not suitable for the extrusion coating process, and will

exhibit excessive edge-weave. As used herein, "neck-in" refers to the reduction in the film width after being extruded. Neck-in is a well known phenomena in the film extrusion art.

EXAMPLE 1

A copolyester resin was made by reacting polyethylene glycol with terephthalic acid, and a small amount (2-4 weight %) of cyclohexenedimethanol. A master blend of the copolyester and calcium carbonate was prepared having a 1:1 ratio. The calcium carbonate had an average particle size of less than about 20 microns, (available from Specialty Minerals, Inc of Adams, MA). This 1:1 master blend was then added to additional unmodified copolyester to form two final blends, the first having a 10 weight % concentration of calcium carbonate and the second having a 20 weight % concentration of calcium carbonate. The two blends were run on the extrusion and laminating line described above. These compositions are then extrusion coated onto bleached Kraft board stock for milk carton or cup stock.

As shown in Fig. 3, the 10% calcium carbonate composition resulted in a dramatic reduction of the neck-in problem. The neck-in exhibited with the 10% composition was about 1.5 inches/side, compared to about 4 inches/side for the same polyester composition without the inorganic filler.

As shown in Fig. 4, coating line speeds were dramatically improved,. The composition containing 10% calcium carbonate was coated at a rate of about twice the line speed of the composition without the filler.

As shown in Fig. 5, the inorganic filler-containing polyester composition had good adhesion to the substrate at a much thinner coating than can be obtained by the polyester composition without the inorganic filler. As used herein "good adhesion" means the cohesive force between the substrate and the polyester was sufficient so that the layers could not be nondestructively separated. Adhesion forces for the materials was measured by the TAPPI method T-439 cm-88 entitled "Determination of Extruded Polymers and the Resulting Adhesion to Porous Substrates" the disclosure of which is incorporated herein by reference. Coatings as thin as 13 microns and having good

adhesion, i.e., having an adhesion level of at least 4, have been achieved with the composition of the present invention.

The same dramatic effects in adhesion, neck-in, and line speeds were also exhibited with the 20% coating. However, at this level of inorganic filler the coating tends to be brittle and is not preferred for this type of application.

EXAMPLES 2-7

The copolyester of Example 1 having about 10 weight % calcium carbonate was extruded. For Examples 2, 3, 5 and 6 a master batch was prepared having 50 weight % calcium carbonate in the copolyester with a final formulation of 90 weight % copolyester and 10 weight % calcium carbonate.

For Example 4 a master batch was prepared having 30 weight % calcium carbonate in DEG-modified copolyester, available from Eastman Chemical Company under the trade name "Eastobond". The final blend having approximately 82 weight % copolyester, 12 weight % Eastobond, and 6 weight % calcium carbonate.

For Example 7 a master batch was prepared having 50 weight % calcium carbonate in a high-IV copolyester (IV = 0.95). The final blend having approximately 80 weight % copolyester, 10 weight % of the high-IV copolyester, and 10 weight % calcium carbonate.

The melt temperature was controlled to approximately 560°F (293°C), and throughput was set at approximately 240 lbs/hr (15 lbs/hr/inch of die width). In the examples, the maximum line speed was determined to be the maximum speed at which the melt curtain was stable, without excessive neck-in or edge weave. The mineral-modified copolyesters were placed in extruders A & B with typical "ramp-up" temperature profiles, targeted for a melt temperature of 560°F (293°C). The mixing block and die temperatures were set at 550°F (288°C). The die opening was set at 76cm, and the line was run at the conditions shown in Table 1 below:

Table 1

Example #	2	3	4	5	6	7
Base Resin	9921	9921	9921/ 19411	9921	9921	9921/ 12822
Mineral Type	none	Onmycarb FT	Hyplex 100	Uncoated $CaCO_3$	talc	Hyplex 100
Max Line Speed, fpm	200	500	300	300	200	600
Neck-In, cm	20	10	15	15	20	7
Minimum thickness for good adhesion	30 microns	15 microns	25 microns	25 microns	30 microns	15 microns

Note: Omnycarb is a 2nd $CaCO_3$,

These examples show the criticality of the choice of mineral. The coated calcium carbonate exhibits superior results with lower neck-in, higher maximum line speed (as determined by the maximum speed before edge weaving became excessive, or greater than 2 cm), and minimum coating thickness required to achieve 100% fiber tearing bond to a bleached board substrate.

The polyester compositions according to the present invention may be coated onto, for instance, paper and paperboard, and also onto other polymers, such as another polyester layer, but is not limited to such substrates. Since there is a dramatic increase in the adhesion properties, numerous other substrates may be used that heretofore were not coatable with a polyester layer. Furthermore, the polyester coatings according to the present invention may have additional coatings thereon, such as silicon-containing coatings, and thus may act, for instance, as a support for silicones onto paper (i.e., the polyester layer is between the silicone layer and the paper layer).

As mentioned, the polyesters used in the composition may be homopolymers or copolymers. Therefore, as used in the appended claims, the term "polyester" includes homopolymers and copolymers, and may contain, for instance, aliphatic and aromatic comonomer units.

Thus, it has been shown that the inclusion of an inorganic filler into a polyester composition provides for an extrusion coating process with improved line speeds and neck-in properties, and a resultant coating that provides good adhesion with very thin coatings of the polyester.

Although the present invention has been described in terms of the presently preferred embodiment, it is to be understood that such disclosure is not to be interpreted as limiting to the invention described herein. No doubt that after reading the disclosure, various alterations and modifications will become apparent to those skilled in the art to which the invention pertains. It is intended that the appended claims be interpreted as covering all such alterations and modifications as fall within the spirit and scope of the invention.

CLAIMS

What is claimed is:

1. A polyester composition comprising a polyester and an inorganic filler, said ~~inorganic filler having an average particle size less than about 50 microns wherein a film~~ of said polyester has a neck-in of less than about 20 centimeters per 76 centimeters of die width.

2. The polyester composition of claim 1 wherein said inorganic filler is calcium carbonate.

3. The polyester composition according to Claim 2, wherein the calcium carbonate is coated.

4. The polyester composition according to Claim 2, wherein the calcium carbonate is not coated.

5. The polyester composition of claim 1 wherein said polyester includes residues of an acid component and a glycol component and at least 80 mole % of said acid component is selected from the group consisting of terephthalic acid, naphthalene-2,6-dicarboxylic acid or an ester thereof and said glycol component is selected from the group consisting of ethylene glycol and polymethylene glycols having 2-10 carbon atoms and selected from the group consisting of trimethylene glycol, tetramethylene glycol, pentamethylene glycol, hexamethylene glycol, and decamethylene glycol.

6. The polyester composition of claim 5 wherein said acid component includes up to about 20 weight % of a modifying dibasic acid having from 2 to 40 carbon atoms.

7. The polyester composition of claim 6 wherein said modifying dibasic acid is selected from the group consisting of isophthalic, succinic, adipic, glutaric, azelaic,

sulfoisophthalic, sebacic, fumaric, cis- or trans- 1,4-cyclohexanedicarboxylic and mixtures thereof.

8. The polyester composition of claim 5 wherein said glycol component comprises from about 97 mole % to about 35 mole % ethylene glycol and from about 1.0 mole % to about 65 mole % 1,4-cyclohexanedimethanol.

9. The polyester composition of claim 8 wherein said glycol component is modified with up to about 20 mole % of a glycol selected from the group consisting of propylene glycol, 1,3-propanediol, neopentyl glycol, 1,4-butanediol, 1,6-hexanediol, 1,4-cyclohexanediol, cis/trans mixtures of 1,4-cyclohexanedimethanol, diethylene glycol, 2,2,4,4-tetramethyl-1,3-cyclobutanediol and mixtures thereof.

10. The polyester composition of claim 2 wherein said calcium carbonate has an average particle size of from about 0.01 microns to about 30 microns.

11. The polyester composition of claim 10 wherein said calcium carbonate has an average particle size of from about 3 microns to about 20 microns.

12. The polyester composition of claim 10 wherein said polyester includes at least 5 weight % of said calcium carbonate.

13. The polyester composition of claim 10 wherein said polyester includes from 5 weight % to about 30 weight % of said calcium carbonate.

14. The polyester composition of claim 10 wherein said polyester includes from about 10 weight % to about 20 weight % of said calcium carbonate.

15. The polyester composition of claim 1 wherein said polyester has a neck-in of less than about 15 centimeters per 76 centimeters of die width.

16. An article of manufacture comprising at least a portion formed from a polyester comprising a polyester and an inorganic filler, said inorganic filler having an average particle size less than about 50 microns wherein a film of said polyester has a neck-in of less than about 20 centimeters per 76 centimeters of die width.

17. The article of claim 16 wherein said filler is coated calcium carbonate having an average particle size of from about 0.01 microns to about 30 microns and said polyester includes at least 5 weight % of said calcium carbonate.

18. The article of claim 16 wherein said filler is coated calcium carbonate having an average particle size of from about 3 microns to about 20 microns and said polyester includes from about 10 weight % to about 20 weight % of said calcium carbonate.

19. The article of manufacture wherein said polyester is coated onto a portion of a substrate.

20. The article of manufacture wherein said substrate is a cellulosic material and wherein said article is selected from the group consisting of packaging containers, and microwaveable food containers and trays.

21. A process for extrusion coating of polyester onto a substrate comprising providing a polyester composition of claim 1 to an extruder; and extruding said polyester composition through at least one die onto a substrate.

22. The process of claim 21 wherein said calcium carbonate is coated and has an average particle size of from about 0.01 microns to about 30 microns.

23. The process of claim 22 wherein said polyester composition includes from about 10 weight % to about 20 weight % of said calcium carbonate.

24. The process of claim 22 wherein said polyester composition includes from about 10 weight % to about 15 weight % of said calcium carbonate.

Figure 1
Side View of Coating Station

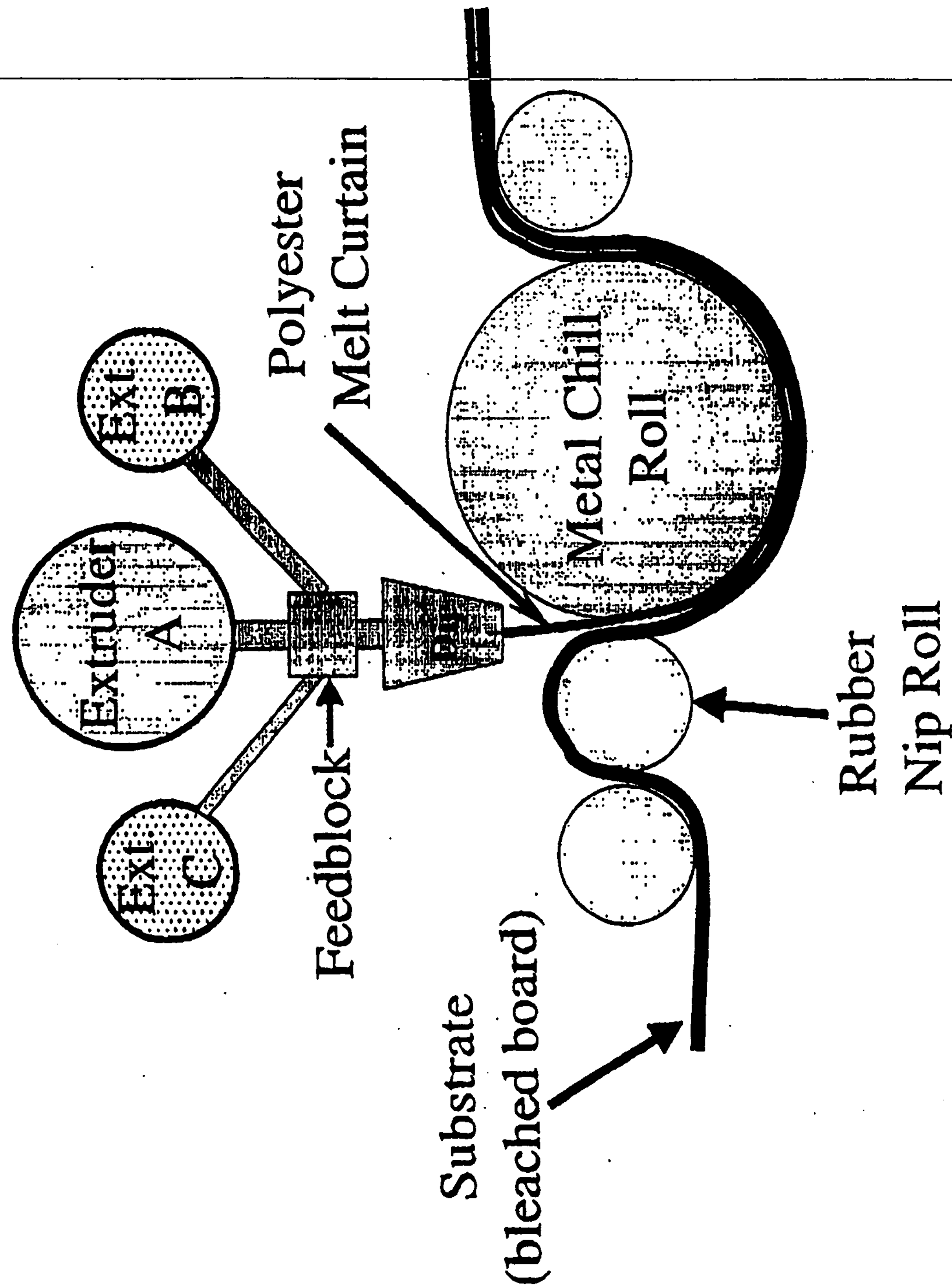
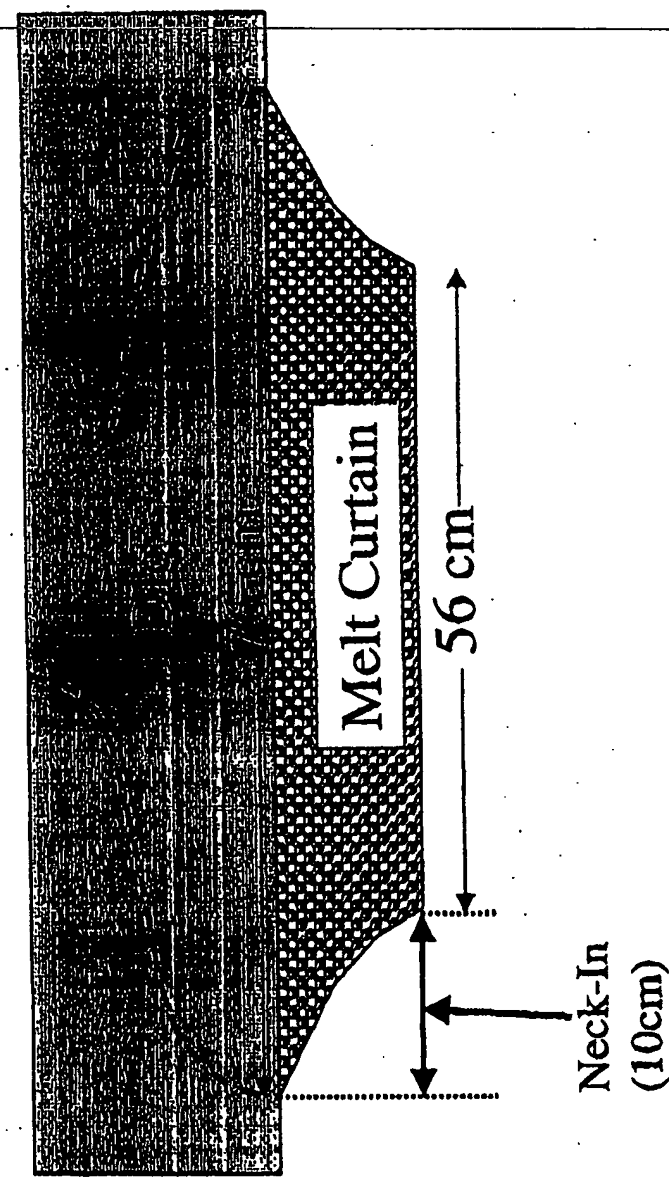


Figure 2
Cross View of Coathanger Die and Polyester Melt Curtain



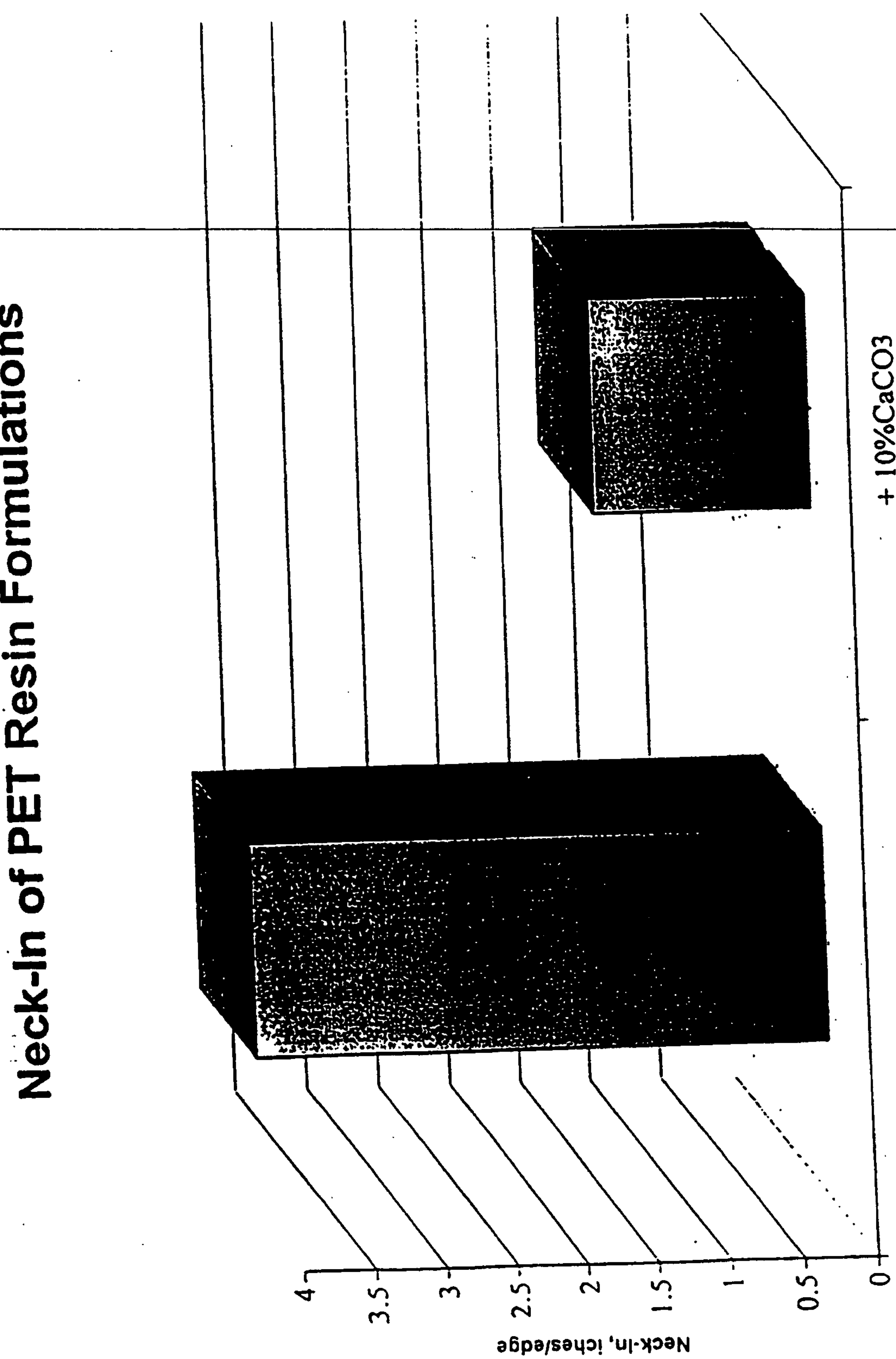

Neck-In of PET Resin Formulations
Neck-In, iches/edge
4
3.5
3
2.5
2
1.5
1
0.5
0
+ 10%CaCO3

Figure 3

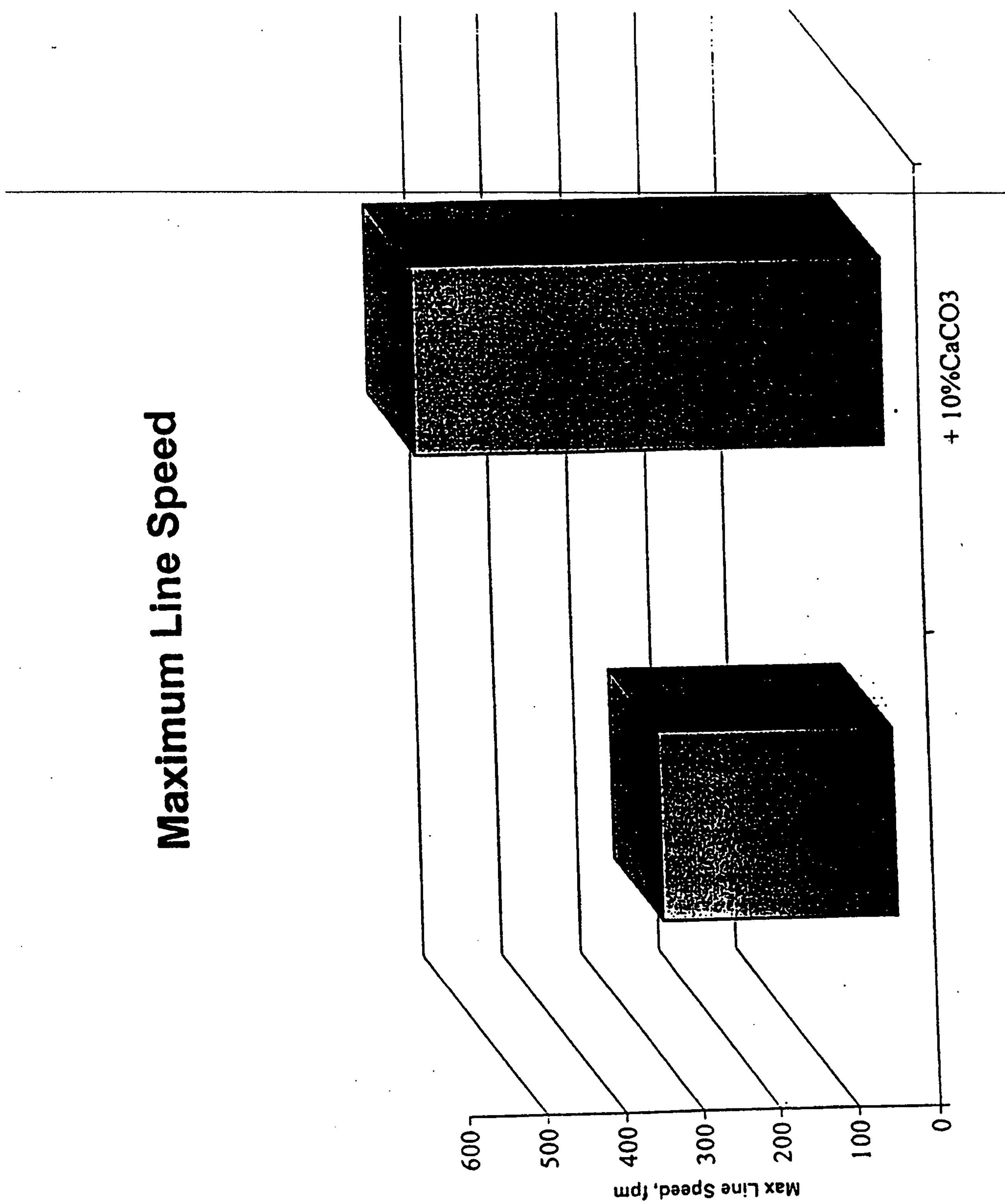

Maximum Line Speed
Max Line Speed, fpm
600
500
400
300
200
100
0
+ 10%CaCO3

Figure 4

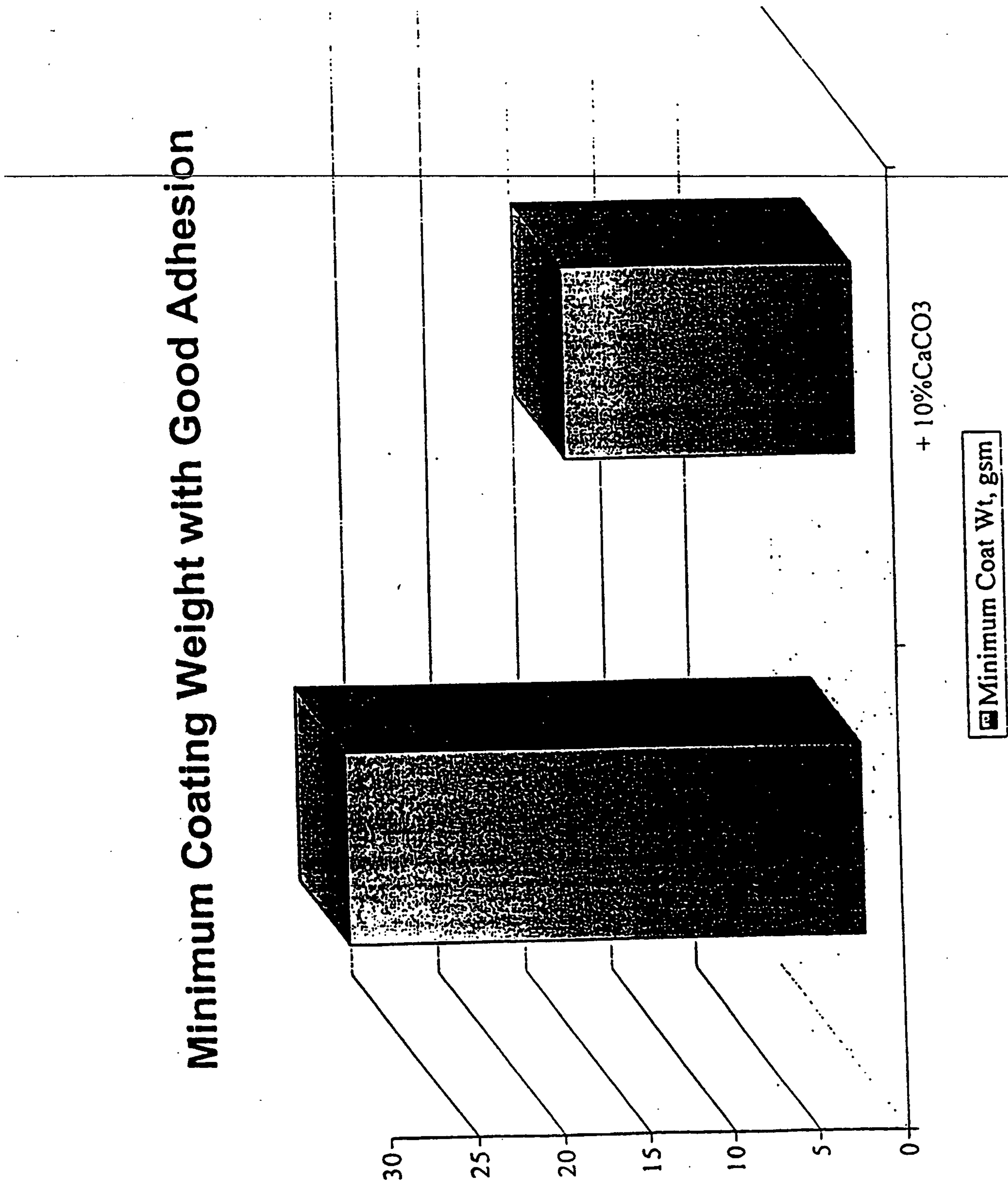

Minimum Coating Weight with Good Adhesion
30
25
20
15
10
5
0
+ 10%CaCO3
Minimum Coat Wt, gsm

Figure 5

INTERNATIONAL SEARCH REPORT

Intern nal Application No
PCT/US 99/01131

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C08K3/00 C08L67/02 C08J5/18

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C08K C08L C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Section Ch, Week 8832 Derwent Publications Ltd., London, GB; Class A23, AN 88-225065 XP002102034 & JP 63 161029 A (TORAY IND INC) , 4 July 1988 see abstract ---	1-15
A	US 5 182 359 A (KANAKA KEIICHI ET AL) 26 January 1993 see column 5, line 32 - line 39 see claim 1 --- -/--	1-15

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
6 May 1999	18/05/1999

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Siemens, T

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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